

"Haruta '394") for the reasons of record. This Final Rejection is traversed for the following reasons.

The Examiner is not persuaded that the term "impregnation" is patentably distinguished over "precipitation," insisting that "impregnation is known in the art to involve precipitation and the two methods are not recognized in the art as mutually exclusive... ." The Examiner is wrong. The terms "impregnation" and "precipitation" are recognized in the art to be mutually exclusive and therefore distinguishable over each other. Several publications in the field of catalysis, relevant pages of which are submitted herewith, provide evidence of the strength of Applicants' position. The Examiner's attention is drawn, for example, to the following quotation from a well-known textbook in heterogeneous catalysis:

"Either of two types of processes, generally termed the *precipitation method* and the *impregnation method*, is commonly used for making catalysts."

*Heterogeneous Catalysis in Practice*, Charles N. Satterfield, Professor of Chemical Engineering MIT, McGraw-Hill Book Company, New York, 1980, pp. 70. (Copy enclosed; underlined emphasis added.)

Satterfield also teaches at page 70 that the precipitation method involves "in the initial stages the mixing of two or more solutions or suspensions of material, causing precipitation..." (Emphasis added.) At page 72 (copy enclosed), Satterfield describes the precipitation method in greater detail, with specifics given as to various anionic precipitants and crystal size. Taking the precipitation method one step further, Satterfield teaches at page 70, last paragraph, "If a carrier is to be incorporated in the final catalyst, the original precipitation is usually carried out in the presence of a suspension of the finely divided support ...." This latter method, which is used by Haruta '394 in precipitating his gold salts/hydroxide onto a titania support, is typically referred to as "deposition precipitation."

In contrast, referring to the impregnation method, Satterfield teaches:

"*Impregnation* is the easiest method of making a catalyst. A carrier, usually porous, is contacted with a solution, usually

aqueous, of one of more suitable metallic compounds. The carrier is then dried, ..."

*Heterogeneous Catalysis in Practice, Ibid., p. 71. (Copy enclosed; underlined emphasis added.)*

At pages 82 and 83 (copies enclosed), the impregnation method is described by Satterfield in greater detail, including the "dipping method" and the "impregnation to incipient wetness" ("dry impregnation") method.

Other artisans skilled in the field also recognize the differences between the impregnation and precipitation methods. See, for example, the following citations:

\*\* Table XIII, wherein the methods of preparing a sulfur-free Ni/SiO<sub>2</sub> catalyst are disclosed as either:

"Various methods of coprecipitation at pH 8.5," *or*

"Dry mix," *or*

"Impregnation at 75°C."

taken from "Structure and Activity of Silica-Supported Nickel Catalysts," by J. W. E. Coenen and B. G. Linsen, in *Physical and Chemical Aspects of Adsorbents and Catalysts*, B. G. Linsen, editor, Academic Press, New York, N.Y., 1970, p.501. (Copy enclosed.)

-----

\*\* "The active precursor can be applied onto the support by:

-adsorption

-impregnation and drying

-precipitation".

J. W. Geus, "Production and Thermal Pretreatment of Supported Catalysts," in *Preparation of Catalysts III*, G. Poncelet, P. Grange, and P. A. Jacobs, editors, Elsevier Science Publishers B.V., Amsterdam, 1983, p. 6. (Copy enclosed.)

-----

In the last reference, a discussion of impregnation follows on pages 6 and 7, whereas a discussion of precipitation and deposition-precipitation follows on page 8 and following. (Pages 6-8 enclosed). In view of all of the above, it is fair for Applicants to assert that impregnation and precipitation are recognized as different techniques in the art of catalyst preparation.

The Examiner asserts that two references (Abel et al., US 5,571,771 and Wang et al., US 5,700,753) illustrate the preparation of an impregnated catalyst made by precipitation; and thus the two techniques are not mutually exclusive. The Examiner misunderstands the technical details of these references. Abel et al. teaches the impregnation of a salt (e.g., palladium, potassium, gold salts) onto a support by contacting a solution of the salt(s) with the support. Abel et al. teaches further that when an impregnated salt is too soluble or contains poisonous anionic species (as in chloride salts), the impregnated salt may be "converted to an insoluble form by precipitation with hydroxides." (Abel et al., Column 3, lines 13-17) The referred-to "precipitation" is actually a surface conversion of an already-impregnated, but too soluble salt into an impregnated and insoluble hydroxide. The overall process involves two steps: impregnation to place the salt onto the surface of the substrate, followed by precipitation with base (fixation as an insoluble hydroxide). Significantly, the impregnation step itself does not involve precipitation. For example, in describing the impregnation step, Abel et al. states, "The solution of the salts should be at a temperature which is high enough *to prevent precipitation of the salts during application to the support.*" (Abel et al., Column 4, lines 21-23; emphasis added.)

The Examiner refers to the Table at Column 8 of Abel et al. as teaching impregnation with precipitation (Table footnote referring to Comparative Example 3b: "#Surface impregnated catalyst in accordance with EP-A-0 519 435 (precipitation with base)"). Comparative Example 3b teaches that a solution of sodium chloropalladate was applied to a support by ultrasound atomizer (an obvious impregnation technique), and then the impregnated support was dried in a hot air stream. *Thereafter*, the impregnated support was treated with sodium hydroxide so as to precipitate palladium and gold hydroxides (i.e., fix an insoluble form onto the support). Note that Abel et al. at this example teaches three distinct steps: impregnation, drying, and thereafter precipitation.

Likewise, Wang et al. describes a two-step method of preparing a catalyst, which involves first impregnation and *thereafter* fixation with a basic alkali metal salt (referred to as precipitation) to secure the impregnated salt to the support. (Wang et

al, at Column 2, lines 30-37) Again, the impregnation step is followed by a distinct precipitation step to form an insoluble and fixated form of the salt.

In view of all of the above, it is submitted that Abel et al. and Wang et al. disclose multi-step catalyst preparation processes, wherein impregnation is then followed by another step of precipitation. The two steps do not occur at the same time and therefore are independent and exclusive of each other.

The Examiner maintains that Haruta '394 discloses gold particles immobilized on a titanium dioxide carrier" which would inherently entail gold particles impregnated onto the carrier." To the contrary, the word "immobilized" *by itself* does not inherently imply *any specific* method of preparation. Moreover, Haruta '394 clearly teaches the details of precipitation and co-precipitation, while making no mention of details characteristic of impregnation. Accordingly, and further in view of the supporting references provided herein by the Applicants, Haruta '394 cannot be interpreted to include inherent teachings to impregnation.

The Examiner maintains that the Applicant "admits that 'impregnation' is known in the art .... and therefore cannot, by itself, be the basis for patentability." The Examiner's argument totally lacks merit. Applicants admit that impregnation is known in the art, as is crystallization, fractional distillation, precipitation, and various other chemical processing techniques. That general knowledge does not preclude obtention of a patent in all of these areas of chemical processing. Did not Haruta '394 obtain a patent on a precipitation method? Patentability lies in the details of the claims. Applicants seek a patent for an impregnation method characterized by specific claimed process steps for preparing a specific catalyst (*impregnation of a gold compound and a reducing agent onto a support, the reducing agent, or the support, or both, comprising titanium, the impregnation being conducted under conditions sufficient to prepare a catalyst comprising gold on a titanium-containing support*). The specific limitations, neither anticipated nor obvious, render Applicants' impregnation process patentable.

The Final Rejection states that the "features upon which Application relies ... are not recited in claim 1." The "features," i.e., limitations, to which the Examiner alludes, comprise the following (as compared with the precipitation method of Haruta

'394), namely: (1) Lower amounts of solvent can be employed. (2) The deposited amount of gold is more reproducible from batch to batch. (3) There is no need to control pH. (4) The time for depositing metal onto the support is quicker. (See Satterfield, which refers to the impregnation method as the "easiest method for preparing catalysts.") These advantages are *inherent characteristics* of the claimed impregnation technique and are *inherently distinguished* over the precipitation technique (which uses larger amounts of solvent, is less reproducible, requires control of pH, and requires longer deposition times). There is no necessity for Applicants to add inherent features to the claims. The limitations that characterize the process and overcome Haruta '394 are already present in the claims.

The Final Rejection states that features upon which Applicants rely, such as "wetting to the point of incipient wetness" are not recited in Claim 1. Applicants see no reason to limit the impregnation method to this preferred, but also more limiting impregnation method. None of the cited references discloses the impregnation process specifically claimed; therefore, the claims are patentable without the additional limitation to incipient wetness. In any case, Claim 28 includes the incipient wetness limitation; therefore at a minimum, this claim should be allowable.

At page 9, the Final Rejection emphasizes a second time that "impregnation is known in the art to involve precipitation and the two methods are not recognized in the art as mutually exclusive." The Final Rejection states, "Claim 1 recites only impregnation and Haruta '394 discloses loading and immobilization onto the support." Applicants vigorously disagree. Impregnation is clearly different from precipitation, as evidenced by the authoritative documents submitted herewith as well as the two references (Abel et al. and Wang et al.) cited by the Examiner. Moreover, Haruta '394's terms of "loading" and "immobilization," taken by themselves, are so general as to be non-descriptive as to the method involved. Taken with its teachings, Haruta '394 unquestionably relates to precipitation and co-precipitation techniques.

The Examiner argues that Claims 22-24 and 27 require promoter metals, which are disclosed by Haruta '394 specifically as adding an alkali compound that would promote precipitation. Indeed, the promoter metals may include alkali compounds, which may be present in the catalyst. Whether the alkali compound

promotes precipitation would depend on the specific alkali compound used, particularly the specific anion employed. Alkali carboxylates generally do not promote precipitation; whereas alkali carbonates or alkali hydroxides may cause precipitation. The impregnation technique, however, requires using a solution of the promoter metal compound(s) or a solution of combined promoter metal compound(s) and gold compound *that does not contain precipitated products*. Once precipitation begins, deposition precipitation would commence; the disadvantages thereof would control; and the deposition would no longer be considered an impregnation.

In view of the above distinctions, it is submitted that Claims 1-9, 18, 20, 22-27, and 29-32 are clearly novel and not anticipated by Haruta '394. It is therefore respectfully requested that the Final Rejection under 35 USC 102(b) be withdrawn.

**B. Concerning the Rejection of Claims 10-14, 16-17, 19, 21, and 28 Under 35 USC 103(a) in view of Haruta '394**

Claims 10-14, 16-17, 19, 21, and 28 stand finally rejected under 35 USC 103(a) as allegedly being unpatentable over Haruta '394, for the reasons of record. This Final Rejection is traversed for the following reasons.

With respect to Claim 10, the Examiner maintains that Haruta '394 suggests the use of reducing agents containing titanium. Although Haruta '394 discloses salts of several carboxylic acids as reducing agents, *titanium salts are not disclosed or suggested*. That Haruta '394 discloses titanium oxide (TiO<sub>2</sub>) as a support is totally immaterial to the point, because a reducing agent and a catalyst support function in distinctly different ways. A support is typically a relatively inert carrier of the catalytic metal. Its purpose typically is to spread-out the catalytic metal on a relatively large surface area. In contrast, a reducing agent is capable of transferring electrons to a reducible atom or compound, herein gold ( $\text{Au}^{+3} \rightarrow \text{Au}^{+<3}$ ). It is crucial to recognize that during reduction, the reducing agent is itself oxidized, since it is losing electrons. The assertion that one skilled in the art would be motivated to select titanium for the carboxylate salt reducing agent, because titania is mentioned as a support material, is far-fetched, because there is no technical nexus between a support and a reducing agent. Moreover, one skilled in the art would know that the **titanium** in titania (TiO<sub>2</sub>) is already in a maximum oxidation state of +4, and cannot give up more electrons (act as a reducing agent) to form a higher oxidation state. (An

oxidation state of greater than +4 is not known for titanium.) All the more reason why the mental connection between a titania support and a titanium reducing agent would not be made by one skilled in the art! Clearly, the Examiner impermissibly employs "hindsight" to construct an argument against the claim. Moreover, even if *arguendo* it were "obvious to try" titanium in the reducing salt, there is no indication or motivation in Haruta '394 that such a combination would be successful in the preparation of the gold-titanium catalyst. "Obvious to try" is not a valid means for defeating a claim.

With regard to Claims 11-14, the Examiner totally misstates Applicants' arguments and misses their point. The Examiner argues that a carboxylic salt "is considered to be an alkyl compound and a carboxylate compound;" and therefore, Haruta '394 by teaching carboxylate salts and titania supports would lead "one of ordinary skill to use titanium as a salt." Carboxylate salts, as noted partially by the Examiner, are typically construed to be alkyl chains terminating in a carboxylate anionic functionality, with the oxygen of the carboxylate functionality being bonded to a metal cation. In contrast, the alkyltitanium and cyclopentadienyl titanium compounds of Claim 12 comprise titanium bonded directly via  $\sigma$ - or  $\pi$ -bonds to alkyl or cyclopentadienyl groups, respectively; i.e., the metal ion (Ti) being directly bonded to carbon. Carboxylate is neither present nor involved in the compounds of Claim 12. Thus, Claim 12 is unquestionably non-obvious, because Haruta '394 is silent regarding the compounds of Claim 12. In addition, Claims 10, 13 and 14, which do embrace the use of titanium carboxylates as reducing agents, are unobvious, because Haruta '394 only mentions titanium in a non-material manner (i.e., supports). It would not be obvious to substitute the support titanium of high oxidation state (+4) into the carboxylate salt that is to be used as a reducing agent. (*Vida supra*.)

With regard to Claim 16, the Examiner again relies on an erroneous connection between precipitation and impregnation methods. Applicants have shown these methods to be patentably distinct. (*Vide supra*)

With regard to Claim 17, the Examiner asserts that "units of measurement are not considered to provide patentable weight and 1/19 is slightly above 5%." This argument is totally without merit. Units of measurement define the parameter measured, and in so doing, can provide patentable weight. The Examiner compares

the gold/titanium atomic ratio of 1/19 (slightly greater than 5 percent Au/Ti) with Applicants' claimed Ti loading of greater than 0.02 to less than 20 weight percent titanium, based on the total weight of the support. The Examiner compares apples (Au/Ti atomic ratio) with oranges (Ti loading on a support); therefore, the argument cannot prevail. Overall, Haruta '394 is silent with regard to titanium loading onto a support, with possibly two inherent exceptions. A skilled artisan might calculate the weight percentage of titanium (Ti) in the disclosed titania support (TiO<sub>2</sub>) to be 59.9 weight percent. (Appendix Z attached.) Alternatively, a skilled artisan might calculate the weight percentage of titanium (Ti) in a mixture of gold and titania, wherein the Au/Ti atomic ratio is 1/19, to be 53.1 weight percent. (Appendix Z attached.) These inherent values are well outside the claimed range of titanium loading.

Claim 19 relates to a titanium-containing reducing agent combined with a variety of specified supports. With regard to Claim 19, the Examiner states "that to practice the invention, one of ordinary skill would need titanium on hand," which is suggested by the disclosure of titania supports. Once again, Applicants disagree, because there is no nexus between reducing agents and titania carriers or supports, and therefore, no motivation to select titanium from the support for the carboxylate salt.

With regard to Claim 28 related to impregnation to the point of incipient wetness, the Examiner asserts that impregnation and precipitation are not mutually exclusive. Applicants have submitted herewith strong evidence to the contrary. Moreover, the Examiner's argument makes no sense. How can a reference to a precipitation method, which involves a completely wet solution technique, suggest a claimed method involving contacting a solid with a solution only to the point of beginning wetness (impregnation to incipient wetness)? The rejection of Claim 28 must be removed.

The Examiner disposes of Haruta 'EP as "not relied upon." The Examiner's argument will fail at the Board of Appeals. All evidence of non-obviousness must be considered when assessing patentability. The Examiner must consider comparative data in determining whether the claimed invention provides unexpected results. *In re Soni*, 34 USPQ2d, 1684, 1687 (Fed. Cir. 1995) Haruta 'EP is highly relevant to the



field of the invention and the very point of this rejection in two ways. Firstly, Haruta 'EP *distinguishes* between the deposition precipitation method and the impregnation method of catalyst preparation. See in Haruta 'EP Examples 1 and 3, drawn to preparation of a catalyst by deposition precipitation and use thereof versus Comparative Example 1, drawn to preparation of the catalyst by impregnation and use thereof. Significantly, Haruta 'EP teaches that the catalyst prepared by impregnation fails to produce any of the desired olefin oxide product in the oxidation of an olefin with oxygen and hydrogen, whereas the catalyst prepared by deposition precipitation is active. Hence, Haruta, himself, teaches to a distinction in catalyst preparation methods. Secondly, it is crucial to note that Haruta 'EP does not employ a reducing agent with the impregnation technique. In contrast, Applicants have now discovered that when a reducing agent is combined with impregnation, then advantageously impregnation --- "the easiest method of preparing catalysts" --- can be employed to prepare gold-titanium catalysts useful for HOPO processes. Applicants' process achieves the desired olefin oxide in high selectivity (>90 mole percent) and at acceptable olefin conversions and desirably low H<sub>2</sub>O/PO molar ratios.

In view of the above, it is submitted that all of Claims 10-14, 16-17, 19, 21, and 28 meet the requirements for non-obviousness. Accordingly, it is respectfully requested that the Final Rejection under 35 USC 103(a) be withdrawn.

**C. Concerning the Final Rejection of Claims 10-17 Under 35 USC 103(a) in View of Haruta '394 taken with Hirose et al.**

Claims 10-17 stand finally rejected under 35 USC 103(a) as allegedly being unpatentable over Haruta '394 in view of Hirose et al., for the reasons of record. This rejection is respectfully traversed for the following reasons.

The Final Rejection attempts to validate Hirose et al. on the grounds that this citation is related to the catalyst arts and discloses a "combination of acetylacetonate of a metal ... with a reducing agent." The argument fails; because, like any field of chemistry, the catalyst arts is a wide-ranging field and notoriously unpredictable. *In re Carleton*, 599 F.2d 1021; 202 U.S.P.Q. 165 (CCPA 1979); *In re Slocombe*, 510 F.2d 1398; 184 U.S.P.Q. 740 (CCPA, 1975); *In re Doumani et al.*, 281 F.2d 215 (CCPA, 1960); 126 USPQ 408. Accordingly, a citation must be close or

pertinent to the field of the invention, and Hirose et al. meets neither criterion. Hirose et al. relates to polyolefin laminates, wherein the polyolefin is produced by a ring-opening polymerization reaction catalyzed by titanyl acetylacetonate. There would be no motivation for a skilled artisan interested in improving a method of preparing gold-titanium hydro-oxidation catalysts to access art related to polyolefin laminates and ring-opening polymerization reactions. Furthermore, the titanyl acetylacetonate catalyst of Hirose et al. may very well be a homogeneous catalyst (soluble in polymerization reaction mixture), whereas the instant claims pertain to preparing a heterogeneous catalyst (gold on an insoluble solid Ti-containing support). A skilled artisan working in the field of heterogeneous chemistry typically does not access prior art in the field of homogeneous chemistry, since homogeneous and heterogeneous catalysts function in such different ways. Accordingly, Hirose et al. has no place in the rejection of claims drawn to an impregnation method of preparing a heterogeneous gold-titanium catalyst for use in hydro-oxidation processes.

Moreover, Applicants even disagree that Hirose et al. discloses what the Examiner says it does. By Applicants' reading, Hirose et al. teaches the titanyl acetylacetonate as being used with "an organoaluminum compound," which is not pertinent to the instant claims, which pertain to the use of organotitanium compounds as reducing agents in the preparation of gold-titanium catalysts for hydro-oxidation processes.

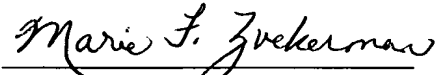
In view of the above, it is submitted that Claims 10-17 are unobvious over Haruta '394 in view of Hirose et al. Accordingly, it is respectfully requested that the Final Rejection of Claims 10-17 under 35 USC 103(a) be withdrawn.

#### D. Conclusions

Applicants trust that the arguments outlined above should persuade the Examiner to reconsider and withdraw the Final Rejection of the instant application.

Applicants believe that Claims 1 to 32 meet all of the standards for patentability. Accordingly, a Notice of Allowance is respectfully requested at the Examiner's earliest convenience.

Respectfully submitted,



Marie F. Zuckerman  
Registration No. 31,315  
Phone: (203) 248-3907

P. O. Box 1967  
Midland, MI 48641-1967

APPENDIX Z - CALCULATIONS OF PERCENTAGE TITANIUM, BY WEIGHT1. Weight percentage of Ti in TiO<sub>2</sub>

$$\begin{array}{rcl} 1 \times \text{Ti atomic weight (47.9 g/mole)} & = & 47.9 \\ 2 \times \text{O atomic weight (16.0 g/mole)} & = & \underline{32.0} \\ \text{Total Weight} & = & 79.9 \text{ g/ mole} \end{array}$$

$$\% \text{ Ti} = (47.9 / 79.9) \times 100\% = 59.9 \%$$

2. Weight percentage of Ti in Mixture of Au and TiO<sub>2</sub> (Au/Ti = 1/19)

$$\begin{array}{rcl} 1 \times \text{Au atomic weight (197.0 g/mole)} & = & 197.0 \\ 19 \times 1 \times \text{Ti atomic weight (47.9 g/mole)} & = & 910.1 \\ 19 \times 2 \times \text{O atomic weight (16.0 g/mole)} & = & \underline{608.0} \\ \text{Total Weight} & = & 1715.1 \text{ g/mole} \end{array}$$

$$\% \text{ Ti} = (910.1 / 1715.1) \times 100\% = 53.1 \%$$